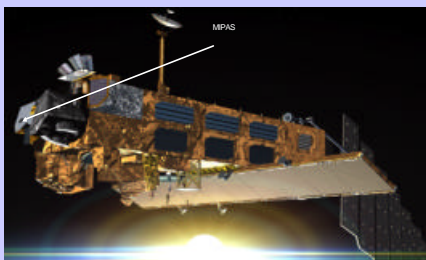


The first two years of the MIPAS/ENVISAT mission: Scientific results related to polar ozone chemistry

Abstract

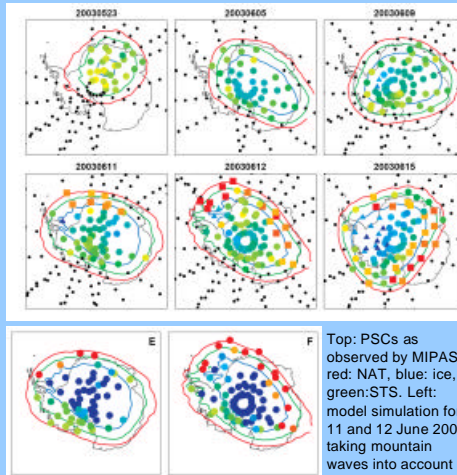
MIPAS on board ENVISAT was launched on 1 March 2002 into a sun-synchronous polar orbit with 98.55° inclination at 800 km altitude. It observes the thermal emission of the atmosphere during day and night by viewing the Earth's limb, covering the globe from pole to pole in an altitude range of 6 to 68 km. From July 2002 to March 2004 MIPAS recorded limb sequences of spectra every 500 km along orbit, resulting in about 1000 limb sequences per day. About 10 % of these spectra have been analyzed at IMK, and have allowed derivation of global fields of temperature, many trace gas distributions, PSCs and clouds.

- The major warming of the Antarctic polar vortex, occurring in September 2002 for the first time since regular observations, has been observed.
- De-nitrification of about 9 ppbv at 475 K was present at the beginning of the major warming and was reduced to 6 ppbv after the vortex split, while the total NO_x deficit remained nearly constant during the event.
- The split led to mixing of NO_x rich air into upper levels of the polar vortex (above 625 K), which resulted in further ozone-loss due to summertime NO_x chemistry.
- Chlorine recovery was observed, with build-up of ClONO₂ as the main pathway, different to former Antarctic winters.
- Global distributions of minor species like HOCl and HO₂NO₂ are currently used to validate in detail chemical transport models of the stratosphere with respect to chlorine, nitrogen, and HO₂ chemistry.
- The chemical composition of PSCs in Antarctic winter 2003 has been identified, and the formation of a NAT belt during this winter has been observed and could be attributed to large-amplitude stratospheric mountain waves over the Antarctic Peninsula.
- In late Arctic winters 2002/2003, high amounts of NO_x, transported from lower latitudes during a major warming, were identified to cause a major part of the accumulated ozone loss due to NO_x summertime chemistry.



Platform: sun-synchronous polar orbiter Envisat
Observation mode: mid-IR limb-emission
Instrument: Michelson interferometer
Spectral resolution: 0.025 cm⁻¹
Horizontal distance between profiles: 550 km
Orbits per day: ~14
Field of view at tangent: 3 (vert) x 30 km (horiz.)

Evolution of NAT PSCs during Antarctic winter 2003

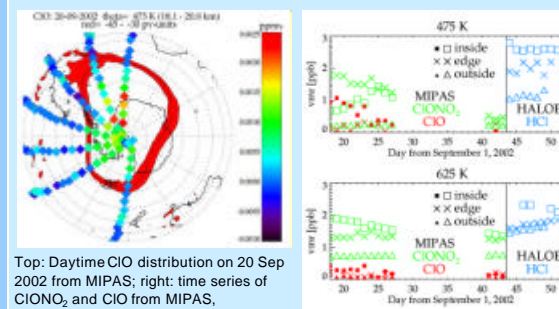


- Chemical composition and microphysical properties (size distribution, volume densities) of PSCs retrieved from spectroscopic signatures.
- Mesoscale microphysical simulations show that sudden onset of NAT PSCs was caused by heterogeneous nucleation on ice in the cooling phase of large amplitude stratospheric mountain waves over the Antarctic Peninsula and the Ellsworth Mountains.
- MIPAS observations of PSCs before this event show no indication for the presence of NAT clouds with volume densities larger than about 0.3 μm³/cm³ and radii smaller than 3 μm, but are consistent with supercooled droplets of ternary H₂SO₄/HNO₃/H₂O solution (STS).
- Simulations indicate that homogeneous surface nucleation rates have to be reduced by three orders of magnitude to comply with the observations.
- The observations suggest a more significant role for mountain waves in Antarctic PSC formation than heretofore appreciated.

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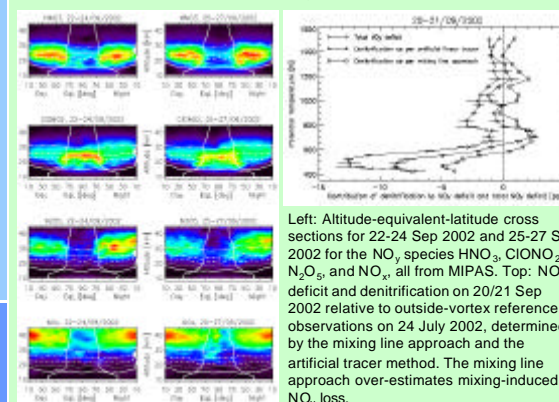
Antarctic major warming 2002: Chlorine recovery



Top: Daytime ClO distribution on 20 Sep 2002 from MIPAS; right: time series of ClONO₂ and ClO from MIPAS, supplemented by HCl from HALOE

- First space-borne observation of ClO by mid-IR limb emission spectroscopy
- Rapid decrease of ClO after disappearance of PSCs on 18 Sep 2002
- Simultaneous increase of ClONO₂ up to 25 Sep indicates recovery of chlorine into ClONO₂, which is unusual for Antarctic winters
- Conversion of ClONO₂ into HCl after 25 Sep, as confirmed by HALOE observations

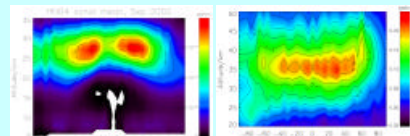
Antarctic major warming Sep 2002: NO_x partitioning and denitrification



Left: Altitude-equivalent-latitude cross sections for 22-24 Sep 2002 and 25-27 Sep 2002 for the NO_x species HNO₃, ClONO₂, N₂O₅, and NO_x, all from MIPAS. Top: NO_x deficit and denitrification on 20/21 Sep 2002 relative to outside-vortex reference observations on 24 July 2002, determined by the mixing line approach and the artificial tracer method. The mixing line approach over-estimates mixing-induced NO_x loss.

- Distributions of NO_x components indicate denitrification at 475 K, re-nitrification due to sedimentation of PSC particles below, and re-nitrification due to NO_x intrusion from lower latitudes above 625 K during the vortex split.
- Outside vortex: NO_x is converted to N₂O₅ when being transported to the pole from low latitudes, ruled by the increased temperatures during the major warming.
- HNO₃ is steadily converted into NO_x by photolysis due to displacement of the vortex to lower latitudes.

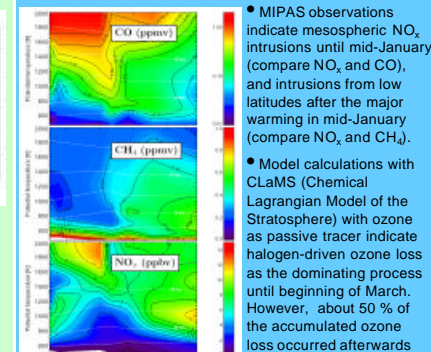
HOCl and HO₂NO₂ global distributions



Left: Averaged global distribution of HO₂NO₂ for Sep 2002. Note a global secondary maximum in the upper troposphere. Right: Averaged global daytime distribution of HOCl. For both, note increased values at high Southern latitudes.

- HOCl altitude range for global retrieval: 20 to 50 km; total precision of individual profiles: 25 % of the vmr peak value; vertical resolution: 9 km. HO₂NO₂ altitude range for global retrieval: 6 to 35 km; total precision of individual profiles: 10 to 15 % of the stratospheric peak vmr and 20 % of the tropospheric peak vmr; vertical resolution: 5 km.
- HOCl couples the catalytic HO₂ and Cl_x cycles, while HO₂NO₂ couples the catalytic NO_x and HO_x cycles. Increase of both during the major warming in September 2002 indicate production of OH and relevance of HO_x catalytic cycle for ozone loss.

Arctic winter 2002/2003: NO_x intrusions



- MIPAS observations indicate mesospheric NO_x intrusions until mid-January (compare NO_x and CO), and intrusions from low latitudes after the major warming in mid-January (compare NO_x and CH₄).
- Model calculations with CLaMS (Chemical Lagrangian Model of the Stratosphere) with ozone as passive tracer indicate halogen-driven ozone loss as the dominating process until beginning of March. However, about 50 % of the accumulated ozone loss occurred afterwards and was driven by NO_x chemistry in ozone-rich air masses transported from low latitudes to the Arctic.
- This kind of ozone depletion could become more important in future if climate change will force a dynamically more active stratosphere with more frequent major warmings.